



PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of : Docket No.: PST6148US1/2159
KARIN BERGSTROM, et al.

Serial No: 10/642,882 : Group Art Unit: 1621

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Title: AN ORTHO ESTER-BASED
SURFACTANT, ITS PREPARATION AND USE

Commissioner for Patents
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DECLARATION UNDER 37 C.F.R. § 1.132

Sir:

I, Per-Erik Hellberg do hereby declare and say that:

1. I am one of the inventors of the above-captioned application;
2. That I have a M.Sc. in Chemical Engineering and a degree of Licentiate of Engineering in Chemistry, both from Chalmers University of Technology, Gothenburg, Sweden
3. That I am a R&D Specialist working with R&D projects related to new types of surfactants and their use in various application areas. I have presently over 16 years of experience in the fields of organic chemistry and surface chemistry.
4. That the following experimentation, prepared by me or under my direct supervision, clearly demonstrate the unexpected superiority of the products of the claimed invention compared to the closest products disclosed by U.S. Patent No. 4,450,087 to Askew et al.

Compounds Tested:

According to U.S. Patent No. 4,450,087 to Askew et al.:

020219HR $(C_4H_9OCH_2CH_2O)_3CH$, 88%, rest $(C_4H_9OCH_2CH_2O)_2CH(OC_2H_5)$
according to 1H - and ^{13}C -NMR *substantially the same as Askew ex. 70*

020222HR2 $[C_4H_9O(CH_2CH_2O)_3]_3CH$, 100%, according to 1H - and ^{13}C -NMR the
compound of *Askew ex. 56*

3xMPEG 350 $[CH_3O(CH_2CH_2O)_{7.2}]_3CH$, 96.5%, rest
 $[CH_3O(CH_2CH_2O)_{7.2}]_2CH(OC_2H_5)$ acc. to 1H - and ^{13}C -NMR

The above compounds are believed to be the closest compounds to the claimed invention disclosed by Askew et al.

According to the present invention.

Products according to Ex. 7 in our case except that the hydrophilic part is ethylene diglycol (EDG) = $C_2H_5CH_2CH_2OCH_2CH_2OH$, plus 0, 3, 7 and 10 ethyleneoxy units (EO) respectively.

GT 6220-02E EDG

GT 6220-05E EDG+3 EO

GT 6220-09E EDG+7 EO

GT 6220-12E EDG+ 10 EO

1) Static Surface Tension

Method

Surface tensions were recorded using a KSV Sigma 70 tensiometer (KSV Instruments LTD, Helsinki, Finland) equipped with a du Noüy ring. Surfactant concentrations were 0.1 % (1 g/L) in pH 8 buffer solution (P-H TAMM Laboratorier AB, Uppsala, Sweden) in order to avoid any hydrolysis during the measurement.

Results - Table 1

Product	Surface Tension (mN/m)
GT 6220-05E	29.7
020222HR2	37.1
3xMPEG 350	59.7

As the data clearly demonstrate, the products of Askew et al. have a substantially lower surface activity.

2) Dynamic Surface Tension

Method

The dynamic surface tension for some of the products earlier described were in this example measured by using a Sensadyne PC500-L Surface Tensiometer (Chem-Dyne Research Corp., Mesa, Az., United States). 0.5 % (5g/L) surfactant solutions were used.

Dissolution in pH 8 buffer solution (P-H TAMM Laboratorier AB, Uppsala, Sweden) in order to avoid any hydrolysis during the measurement.

The surface tension was measured as a function of time. The solutions were continuously stirred except during the measurements. With the method used*, the surface tension is measured by creation of nitrogen bubbles in the bulk phase. The process is dynamic, and the surface tension measured depends on the surface age of the bubble. A higher surface age normally means a lower surface tension until the creation time is so long that steady-state conditions are reached. The surface age at which this takes place is individual for each surfactant.

* The method is described in e.g. *U. Teipel and N. Aksel, Tenside Surf. Det. 37 (2000) 5, 297-308* and references therein.

Results

Table 2

Product	Surface age (s)	Surface Tension (mN/m)	Surface age (s)	Surface Tension (mN/m)
GT 6220-05E	0.65	31.6	2.34	29.7
GT 6220-09E	0.69	31.0	2.46	29.4
GT 6220-12E	0.64	34.8	2.49	29.8
020222HR2	0.62	35.7	2.89	36.4
3xMPEG 350	0.67	63.5	2.70	63.2

Although the surface tension for 020222HR2 of Askew et al. is fairly low, the surface tension of the products of the claimed invention are substantially lower demonstrating unexpected superiority for the claimed products.

3) Dynamic Wetting

Method

Measurements were carried out by depositing droplets on a hydrophobic, polymeric material (Parafilm PM-922, American Can Company) using a First Ten Ångstroms FTÅ 200 instrument (Portsmouth, VA, USA) equipped with a video camera and an image analyzing unit. The surfactants were formulated as follows prior to measurement: Surfactant 5.0 % w/w, tetrasodium pyrophosphate (TKPP) 6.0 % w/w, sodium metasilicate pentahydrate 4.0 % w/w and balance distilled, de-ionized water (18 MΩ/cm). The formulations were diluted 1:20 with distilled, de-ionized water before measurement.

This is similar to ex. 11 in the present application, although surfactant concentration is lower in this case.

Results

Table 3

Product	Contact Angle (°) after		
	10 s	40 s	70 s
GT 6220-05E	49,2	45,2	42,6
GT 6220-09E	47,4	43,1	40,1
GT 6220-12E	46,4	40,3	38,7
020222HR2	66,9	67,0	66,1
3xMPEG 350	89,0	88,3	87,6

In addition to the much higher contact angle (inferior wetting) for the products of Askew et al., the lack of surfactancy is also demonstrated by the flat curve (no reduction in contact angle with time).

4) Wetting according to Draves

Method

The wetting capability is determined by examination of the time consumed for wetting a skein of yarn when the concentration of the solution is 1 g surfactant/liter. The skein is double-folded and equipped with a hook and a 40 g sinker of lead. Time from immersion until the lower part of the hook is on level with the upper part of the sinker is measured.

Ref. ASTM 02281

Results

Table 4

Product	Wetting time (s)
GT 6220-05E	41
GT 6220-09E	35
GT 6220-12E	55
020222HR2	> 600
3xMPEG 350	> 600

According to wetting method ASTM 02281 the products of Askew have virtually no wetting capacity, while the products according to the present invention do demonstrate significant and unexpectedly superior wetting capacity.

5) Emulsification

Method

The emulsification was carried out by mixing in a 22 mL vial using an Ultra-Turrax at 8000 rpm for 60 seconds. Emulsion status was then checked with the naked eye at regular time intervals after end of emulsification. Clearly separated oil- or waterphases (in mm) were noted. Rest of height was emulsion phase. Oil phase and volumes were varied but aqueous phase was in all cases pH 8 buffer solution(P-H TAMM Laboratorier AB, Uppsala, Sweden) in order to avoid any hydrolysis during the measurement.

Results

Oil = decane (2g) + 1 dr Sudan Red, aq. phase = 6g, surfactant = 0.40g
Excess aq phase.

Time (min)	GT 6220-05E		GT 6220-09E		GT 6220-12E		020222HR2		3xMPEG 350	
	Sep. oil	Sep. water								
0,5	0	0	0	0	0	0	8	17	5	5
1	0	0	0	0	0	0	9	19	5	9
1,5	0	0	0	0	0	0			6	17
2	0	0	0	0	0	0			8	19
3	0	0	0	0	0	0			9	19
4	0	0	0	0	0	1				
8	1	1	0	0	0	3				
10	1	1	0	2	0	3				
15	2	2	0	2	0	6				

Grey shade = complete separation

The data clearly demonstrate the superior emulsification properties of the compounds according to the claimed invention compared to the compounds of Askew et al. over time. More specifically, there was complete separation of Askew et al.'s compounds after 1 and three minutes, respectively, while the compounds of the claimed invention substantially better emulsification for as long as 15 minutes.

Summary/Conclusion

In summary, several of the compounds disclosed by Askew et al. that are believed to be the closest to the presently claimed compounds were tested in side by side comparisons to the claimed compounds. They were compared in

1. Static Surface Tension,
2. Dynamic Surface Tension,
3. Dynamic Wetting,
4. Wetting according to Draves (ASTM 02281), and
5. Emulsification.

Analysis of the test results and data clearly leads one to conclude that the compounds of the present invention are unexpectedly superior in the parameters tested, i.e., they are superior surfactants, while the compounds of Askew et al. do not have good surface activity.

The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under Section 1001 Title 18 of the United States Codes and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Date: March 11, 2004


PER-ERIK HELLBERG